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Abstract

In a long wavelength regime, the effective properties of particulate composites, including nanocomposites, may be estimated using one of various homogenization formalisms, such as the Bruggeman and Maxwell Garnett formalisms, and the approach of the strong–property–fluctuation theory (SPFT). In the conventional implementations of these formalisms, the constituent particles are treated as point–like scattering centres. However, extended formalisms have been established — which involve integral formulations — that take account of the spatial extent of the constituent particles. In particular, the extended second–order SPFT takes account of both the size of the constituent particles and their statistical distributions. We derive explicit representations of the extended second–order SPFT appropriate to isotropic chiral and uniaxial dielectric homogenized composite mediums. These results may also be employed in extended versions of the Bruggeman and Maxwell Garnett formalisms.

keywords: strong–property–fluctuation theory, depolarization dyadic, Bruggeman formalism, Maxwell Garnett formalism

1 Introduction

In the present era of exotic composite materials with nanoscale architectures, there is a pressing need for accurate theoretical tools to predict their electromagnetic properties [1]. While this generally poses a formidable challenge to theoreticians, matters can be simplified considerably provided that wavelengths are sufficiently long relative to the length scales of the particles from which the composite is assembled — the composite may then be regarded as being effectively homogeneous. Various well established formalisms are available to estimate the constitutive parameters of such homogenized composite mediums (HCMs) [2]. One of the most sophisticated — and one which has also gained prominence lately in studies of HCMs as metamaterials [3] — is based on the strong–property–fluctuation theory (SPFT) [4].

The origins of the SPFT lie in wave propagation studies pertaining to continuous random mediums [5, 6], but it was later adapted to estimate the constitutive parameters of HCMs [4, 7, 8, 9, 10]. Unlike other more commonly used approaches to homogenization, such as the Bruggeman and Maxwell Garnett formalisms [11], the SPFT can accommodate a comprehensive description of the distributional statistics of the constituent particles. The SPFT provides an estimate of the HCM constitutive parameters via a recursive scheme, based on an apt ambient medium described

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by the Bruggeman formalism. Usually the recursive scheme is truncated at the second–order level, wherein the spatial distribution statistics of the constituent particles are described in terms of a two–point covariance function and its associated correlation length. In a recent development, the *extended* SPFT was established which takes into account the spatial extent of the constituent particles [12, 13]. Similarly extended versions of the Maxwell Garnett and Bruggeman formalisms have also been developed [14, 15, 16, 17, 18], but these apply only to isotropic HCMs (whereas the extended SPFT is available for bianisotropic HCMs [13]) and involve simplistic descriptions of the constituent particle distributions.

A drawback with the SPFT approach to homogenization is that its implementation can be an involved process, generally requiring numerical methods. Explicit expressions (i.e., ones not expressed in terms of integrals) are not available for SPFT estimates of HCM constitutive parameters, with the exception of certain isotropic dielectric homogenization scenarios [4]. We address this issue in the following by deriving explicit expressions for the extended SPFT appropriate to isotropic chiral and uniaxial dielectric HCMs. The expressions derived for depolarization dyadics can also be utilized in extended versions of the Bruggeman formalism and the Maxwell Garnett formalism.

In the following, vector quantities are underlined. Double underlining and normal (bold) face signify a 3×3 (6×6) dyadic. The determinant, inverse and transpose of a dyadic $\underline{\underline{M}}$ are denoted by $\det\left[\underline{\underline{M}}\right]$, $\underline{\underline{M}}^{-1}$ and $\underline{\underline{M}}^{T}$, respectively. The 3×3 (6×6) identity dyadic is represented by $\underline{\underline{I}}$ ($\underline{\underline{\mathbf{I}}}$); and the 3×3 (6×6) null dyadic is represented by $\underline{\underline{0}}$ ($\underline{\underline{\mathbf{0}}}$). Angular frequency is denoted by ω; the permeability of free space is μ_0; and $i = \sqrt{-1}$. The homogeneous bianisotropic medium specified by the (frequency–domain) Tellegen constitutive relations [19]

$$\left.\begin{array}{l} \underline{D}(\underline{r}) = \underline{\underline{\epsilon}}_\ell \cdot \underline{E}(\underline{r}) + \underline{\underline{\xi}}_\ell \cdot \underline{H}(\underline{r}) \\ \underline{B}(\underline{r}) = \underline{\underline{\zeta}}_\ell \cdot \underline{E}(\underline{r}) + \underline{\underline{\mu}}_\ell \cdot \underline{H}(\underline{r}) \end{array}\right\} \tag{1}$$

is compactly characterized by its 6×6 constitutive dyadic

$$\underline{\underline{\mathbf{K}}}_\ell = \left[\begin{array}{cc} \underline{\underline{\epsilon}}_\ell & \underline{\underline{\xi}}_\ell \\ \underline{\underline{\zeta}}_\ell & \underline{\underline{\mu}}_\ell \end{array}\right], \tag{2}$$

which subsumes the four 3×3 constitutive dyadics $\underline{\underline{\epsilon}}_\ell$, $\underline{\underline{\xi}}_\ell$, $\underline{\underline{\zeta}}_\ell$ and $\underline{\underline{\mu}}_\ell$. Subscripts are used to identify the particular medium that the constitutive dyadics describe.

2 Analysis

Many approaches to homogenization, including those of the SPFT and the Bruggeman and Maxwell Garnett formalisms, rely on depolarization dyadics to represent the scattering responses of the constituent particles. General integral formulations of depolarization dyadics are presented in §2.1; and we show how these are incorporated into the SPFT in §2.2. In §2.3 the main results of this communication are presented as explicit representations of the extended SPFT for isotropic chiral HCMs and uniaxial dielectric HCMs.

2.1 Depolarization region

Let us consider a homogeneous spherical particle of radius η, embedded in a homogeneous ambient medium characterized by the 6×6 constitutive dyadic $\underline{\underline{\mathbf{K}}}_{\text{amb}}$. Provided that:

(i) current density distributions induced within the particle are uniform throughout its volume, and

(ii) the particle is small relative to electromagnetic wavelengths,

the particle's scattering response is captured by the depolarization dyadic [20, 21]

$$\underline{\underline{\mathbf{D}}}(\eta) = \int_{|\underline{r}|<\eta} \underline{\underline{\mathbf{G}}}_{\rm amb}(\underline{r})\; d^3\underline{r}. \tag{3}$$

Herein, $\underline{\underline{\mathbf{G}}}_{\rm amb}(\underline{r})$ is the 6×6 dyadic Green function of the ambient medium. While explicit representations of $\underline{\underline{\mathbf{G}}}_{\rm amb}(\underline{r})$ are not generally available for anisotropic and bianisotropic ambient mediums [22], its Fourier transform, namely

$$\underline{\underline{\tilde{\mathbf{G}}}}_{\rm amb}(\underline{q}) = \int_{\underline{r}} \underline{\underline{\mathbf{G}}}_{\rm amb}(\underline{r})\; \exp(-i\underline{q}\,\bullet\,\underline{r})\; d^3\underline{r}\,, \tag{4}$$

is expressible as [23]

$$\underline{\underline{\tilde{\mathbf{G}}}}_{\rm amb}(\underline{q}) = \frac{1}{i\omega}\; \left[\underline{\underline{\tilde{\mathbf{A}}}}_{\rm amb}(\underline{q})\right]^{-1}, \tag{5}$$

where

$$\underline{\underline{\tilde{\mathbf{A}}}}_{\rm amb}(\underline{q}) = \left[\begin{array}{cc} \underline{\underline{0}} & (\underline{q}/\omega)\times\underline{\underline{I}} \\ -(\underline{q}/\omega)\times\underline{\underline{I}} & \underline{\underline{0}} \end{array}\right] + \underline{\underline{\mathbf{K}}}_{\rm amb}. \tag{6}$$

Exploiting the spectral representation (4), the depolarization dyadic is given by [20, 23]

$$\underline{\underline{\mathbf{D}}}(\eta) \;=\; \frac{\eta}{2\pi^2}\int_{\underline{q}} \frac{1}{q^2}\left[\frac{\sin(q\eta)}{q\eta} - \cos(q\eta)\right] \underline{\underline{\tilde{\mathbf{G}}}}_{\rm amb}(\underline{q})\; d^3\underline{q}, \tag{7}$$

with $q^2 = \underline{q}\,\bullet\,\underline{q}$.

In order to accommodate a depolarization region of nonzero volume, the depolarization dyadic may be considered as the sum [12, 13]

$$\underline{\underline{\mathbf{D}}}(\eta) = \underline{\underline{\mathbf{D}}}^0 + \underline{\underline{\mathbf{D}}}^+(\eta), \tag{8}$$

wherein

$$\underline{\underline{\mathbf{D}}}^0 = \frac{\eta}{2\pi^2}\int_{\underline{q}} \frac{1}{q^2}\left[\frac{\sin(q\eta)}{q\eta} - \cos(q\eta)\right] \underline{\underline{\tilde{\mathbf{G}}}}^{\infty}_{\rm amb}(\hat{\underline{q}})\; d^3\underline{q}\,, \tag{9}$$

$$\underline{\underline{\mathbf{D}}}^+(\eta) = \frac{\eta}{2\pi^2}\int_{\underline{q}} \frac{1}{q^2}\left[\frac{\sin(q\eta)}{q\eta} - \cos(q\eta)\right] \underline{\underline{\tilde{\mathbf{G}}}}^{+}_{\rm amb}(\underline{q})\; d^3\underline{q}\,, \tag{10}$$

with

$$\underline{\underline{\tilde{\mathbf{G}}}}^{\infty}_{\rm amb}(\hat{\underline{q}}) \;=\; \lim_{q\to\infty}\; \underline{\underline{\tilde{\mathbf{G}}}}_{\rm amb}(\underline{q}), \tag{11}$$

$$\underline{\underline{\tilde{\mathbf{G}}}}^{+}_{\rm amb}(\underline{q}) \;=\; \underline{\underline{\tilde{\mathbf{G}}}}_{\rm amb}(\underline{q}) - \underline{\underline{\tilde{\mathbf{G}}}}^{\infty}_{\rm amb}(\hat{\underline{q}}), \tag{12}$$

and the unit vector $\hat{\underline{q}} = \underline{q}/q = (\sin\theta\cos\phi,\; \sin\theta\sin\phi,\; \cos\theta)$. The dyadic $\underline{\underline{\mathbf{D}}}^0$ represents the depolarization contribution arising from the vanishingly small spherical region in the limit $\eta \to 0$,

whereas the dyadic $\underline{\underline{\mathbf{D}}}^{+}(\eta)$ provides the depolarization contribution arising from the spherical region of nonzero volume. It is widespread practice in homogenization studies to neglect $\underline{\underline{\mathbf{D}}}^{+}(\eta)$ and simply take $\underline{\underline{\mathbf{D}}}^{0}$ as the depolarization dyadic [24]. However, the importance of the spatial extent of depolarization regions has been underlined in studies of isotropic [25, 26, 14, 16, 17], anisotropic [12] and bianisotropic [13] HCMs.

The integral representation of $\underline{\underline{\mathbf{D}}}^{0}$ has been extensively studied [24]. The volume integral in eq. (9) simplifies to the η–independent surface integral [20, 23]

$$\underline{\underline{\mathbf{D}}}^{0} = \frac{1}{4\pi} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \underline{\underline{\tilde{\mathbf{G}}}}^{\infty}_{\text{amb}}(\hat{\underline{q}}) \ \sin\theta \ d\theta \ d\phi, \tag{13}$$

which is easily evaluated for isotropic dielectric–magnetic and isotropic chiral mediums [19]. Explicit evaluations for uniaxial dielectric mediums have been presented in terms of hyperbolic functions [20], whereas an elliptic function representation is available for biaxial dielectric mediums [27].

The depolarization contribution given by $\underline{\underline{\mathbf{D}}}^{+}(\eta)$ has been computed in various numerical studies [12, 13], but hitherto no explicit evaluations of the volume integral in eq. (10) have been reported for ambient mediums other than isotropic dielectric ambient mediums [4]. A notable simplification arises in the case of Lorentz–reciprocal ambient mediums (i.e., ambient mediums which satisfy $\underline{\underline{\epsilon}}_{\text{amb}} = \underline{\underline{\epsilon}}^{T}_{\text{amb}}$, $\underline{\underline{\xi}}_{\text{amb}} = -\underline{\underline{\zeta}}^{T}_{\text{amb}}$, and $\underline{\underline{\mu}}_{\text{amb}} = \underline{\underline{\mu}}^{T}_{\text{amb}}$ [28]). Therein $\underline{\underline{\mathbf{D}}}^{+}(\eta)$ has the surface integral representation [10]

$$\begin{aligned}
\underline{\underline{\mathbf{D}}}^{+}(\eta) \ = \ & \frac{\omega^4}{4\pi} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \frac{1}{b_{\,\text{amb}}(\theta,\phi)} \left[\frac{1}{\kappa_+ - \kappa_-} \left(\frac{e^{i\eta q}}{2q^2} \, (1 - i\eta q) \left\{ \det\left[\underline{\underline{\tilde{\mathbf{A}}}}_{\text{amb}}(\underline{q}) \right] \underline{\underline{\tilde{\mathbf{G}}}}^{+}_{\text{amb}}(\underline{q}) \right. \right. \right. \\
& \left. \left. \left. + \det\left[\underline{\underline{\tilde{\mathbf{A}}}}_{\text{amb}}(-\underline{q}) \right] \underline{\underline{\tilde{\mathbf{G}}}}^{+}_{\text{amb}}(-\underline{q}) \right\} \right)_{q=\sqrt{\kappa_-}}^{q=\sqrt{\kappa_+}} + \frac{\det\left[\underline{\underline{\tilde{\mathbf{A}}}}_{\text{amb}}(\underline{0}) \right]}{\kappa_+ \kappa_-} \underline{\underline{\tilde{\mathbf{G}}}}^{+}_{\text{amb}}(\underline{0}) \right] \sin\theta \ d\theta \ d\phi,
\end{aligned} \tag{14}$$

with $\kappa_\pm$ being the q^2 roots of $\det\left[\underline{\underline{\tilde{\mathbf{A}}}}_{\text{amb}}(\underline{q}) \right] = 0$ and the scalar function

$$b_{\,\text{amb}}(\theta,\phi) = \left(\hat{\underline{q}} \cdot \underline{\underline{\epsilon}}_{\text{amb}} \cdot \hat{\underline{q}} \right) \left(\hat{\underline{q}} \cdot \underline{\underline{\mu}}_{\text{amb}} \cdot \hat{\underline{q}} \right) + \left(\hat{\underline{q}} \cdot \underline{\underline{\xi}}_{\text{amb}} \cdot \hat{\underline{q}} \right)^2. \tag{15}$$

Evaluations of the surface integral on the right side of eq. (14) are provided in §2.3.1 and §2.3.2 for isotropic chiral ambient mediums and uniaxial dielectric ambient mediums, respectively.

2.2 Homogenization

Depolarization dyadics play a central role in formalisms employed to estimate the constitutive parameters of HCMs [4]. Here we focus on the approach provided by the extended SPFT [12, 13]. We concentrate on the homogenization of a two–phase composite wherein the two constituent phases, labelled as a and b, comprise spherical particles of average radius η. The constituent phase a occupies the region V_{a} whereas constituent phase b occupies the region V_{b}, The constituent phases are randomly mixed with their distributional statistics being described in terms of moments of the characteristic functions

$$\Phi_\ell(\underline{r}) = \begin{cases} 1, & \underline{r} \in V_\ell, \\ 0, & \underline{r} \notin V_\ell, \end{cases} \qquad (\ell = \text{a,b}). \tag{16}$$

The volume fraction of phase ℓ, namely f_ℓ , is given by the first statistical moment of Φ_ℓ ; i.e., $\langle\, \Phi_\ell(\underline{r})\,\rangle = f_\ell$. We have $\langle\, \Phi_{\rm a}(\underline{r})\,\rangle + \langle\, \Phi_{\rm b}(\underline{r})\,\rangle = 1$. For the second statistical moment of Φ_ℓ, the physically–motivated covariance form [29]

$$\langle\, \Phi_\ell(\underline{r})\, \Phi_\ell(\underline{r}')\,\rangle = \begin{cases} \langle\, \Phi_\ell(\underline{r})\,\rangle \langle \Phi_\ell(\underline{r}')\,\rangle\,, & |\underline{r}-\underline{r}'| > L, \\ \\ \langle\, \Phi_\ell(\underline{r})\,\rangle\,, & |\underline{r}-\underline{r}'| \leq L, \end{cases} \qquad (\ell = \text{a,b}), \tag{17}$$

is commonly implemented. Herein, the correlation length L is required to be much smaller than the electromagnetic wavelengths, but much larger than the constituent particle radius η. Parenthetically, the specific form of the covariance function has only a secondary influence on SPFT estimates of HCM constitutive parameters, across a range of physically–plausible covariance functions [30].

The nth–order SPFT estimate of the HCM constitutive dyadic, namely $\underline{\underline{\mathbf{K}}}_{\rm HCM}^{[n]}$, derives from the recursive refinement of an ambient homogeneous medium, characterized by the constitutive dyadic $\underline{\underline{\mathbf{K}}}_{\rm amb}$. At lowest order (i.e., zeroth and first order), the SPFT estimate of the HCM constitutive dyadic is identical to that of the ambient medium [10]; i.e.,

$$\underline{\underline{\mathbf{K}}}_{\rm HCM}^{[0]} = \underline{\underline{\mathbf{K}}}_{\rm HCM}^{[1]} = \underline{\underline{\mathbf{K}}}_{\rm amb}. \tag{18}$$

Furthermore, $\underline{\underline{\mathbf{K}}}_{\rm amb}$ is delivered by solving the nonlinear equations

$$f_{\rm a}\, \underline{\underline{\boldsymbol{\chi}}}_{\rm a}(\eta) + f_{\rm b}\, \underline{\underline{\boldsymbol{\chi}}}_{\rm b}(\eta) = \underline{\underline{\mathbf{0}}}, \tag{19}$$

wherein the polarizability density dyadics

$$\underline{\underline{\boldsymbol{\chi}}}_{\ell}(\eta) = -i\omega \left(\underline{\underline{\mathbf{K}}}_{\ell} - \underline{\underline{\mathbf{K}}}_{\rm amb} \right) \cdot \left[\underline{\underline{\mathbf{I}}} + i\omega \underline{\underline{\mathbf{D}}}(\eta) \cdot \left(\underline{\underline{\mathbf{K}}}_{\ell} - \underline{\underline{\mathbf{K}}}_{\rm amb} \right) \right]^{-1}, \qquad (\ell = \text{a,b}), \tag{20}$$

and $\underline{\underline{\mathbf{K}}}_{\ell}$ ($\ell = $ a,b) are the constitutive dyadics of constituent phases a and b.

For a broad range of bianisotropic HCMs, the SPFT (including the extended SPFT [31]) converges at the second-order level[2] [32]. The second–order SPFT estimate of the HCM constitutive dyadic is [10]

$$\underline{\underline{\mathbf{K}}}_{\rm HCM}^{[2]} = \underline{\underline{\mathbf{K}}}_{\rm amb} - \frac{1}{i\omega} \left[\underline{\underline{\mathbf{I}}} + \underline{\underline{\boldsymbol{\Sigma}}}^{[2]}(\eta, L) \cdot \underline{\underline{\mathbf{D}}}(\eta) \right]^{-1} \cdot \underline{\underline{\boldsymbol{\Sigma}}}^{[2]}(\eta, L), \tag{21}$$

with the mass operator dyadic term [5] — corresponding to the covariance function (17) — being

$$\underline{\underline{\boldsymbol{\Sigma}}}^{[2]}(\eta, L) = f_{\rm a} f_{\rm b} \left[\underline{\underline{\boldsymbol{\chi}}}_{\rm a}(\eta) - \underline{\underline{\boldsymbol{\chi}}}_{\rm b}(\eta) \right] \cdot \underline{\underline{\mathbf{D}}}^{+}(L) \cdot \left[\underline{\underline{\boldsymbol{\chi}}}_{\rm a}(\eta) - \underline{\underline{\boldsymbol{\chi}}}_{\rm b}(\eta) \right]. \tag{22}$$

Thus, within the extended second–order SPFT, the estimate of the HCM constitutive dyadic depends on two length scales: the constituent particle size η via the depolarization dyadic and the correlation length L via the mass operator.

2.3 Explicit representations

We now turn to the evaluation of the surface integral which delivers $\underline{\underline{\mathbf{D}}}^{+}$. This term crops up as $\underline{\underline{\mathbf{D}}}^{+}(\eta)$ in the extended depolarization dyadic representation (8) and as $\underline{\underline{\mathbf{D}}}^{+}(L)$ in the mass operator term (22) which yields the second–order SPFT estimate of the HCM constitutive dyadic. A key

[2]The second–order SPFT approximation is also known as the bilocal approximation.

step in the evaluation of the integral on the right side of eq. (14) is the approximation of the $e^{i\eta q}\,(1-i\eta q)$ term in the integrand by its asymptotic expansion $1+(\eta q)^2/2+i(\eta q)^3/3$, which is permissible since $\eta\sqrt{\kappa_\pm}\ll 1$ in the long wavelength regime. The evaluation of $\underline{\underline{\mathbf{D}}}^+(L)$ is isomorphic to that of $\underline{\underline{\mathbf{D}}}^+(\eta)$ since here we similarly have $L\sqrt{\kappa_\pm}\ll 1$ in the long wavelength regime. In the following we consider two types of ambient medium: an isotropic chiral ambient medium in §2.3.1 and a uniaxial dielectric ambient medium in §2.3.2.

2.3.1 Isotropic chiral ambient medium

Suppose that the ambient medium is an isotropic chiral medium. Its constitutive dyadic has the form [33]

$$\underline{\underline{\mathbf{K}}}_{\rm amb} = \left[\begin{array}{cc} \epsilon_{\rm amb}\,\underline{\underline{I}} & \xi_{\rm amb}\,\underline{\underline{I}} \\ -\xi_{\rm amb}\,\underline{\underline{I}} & \mu_{\rm amb}\,\underline{\underline{I}} \end{array}\right]. \tag{23}$$

The integral on the right side of eq. (14) may then be evaluated to give

$$\begin{aligned} \underline{\underline{\mathbf{D}}}^+(\eta) \;=\; & \frac{i\omega\eta^2}{3}\Bigg(\left[\begin{array}{cc} \mu_{\rm amb}\,\underline{\underline{I}} & \xi_{\rm amb}\,\underline{\underline{I}} \\ -\xi_{\rm amb}\,\underline{\underline{I}} & \epsilon_{\rm amb}\,\underline{\underline{I}} \end{array}\right] \\ & + i\frac{2\omega\eta}{3}\left[\begin{array}{cc} \sqrt{\dfrac{\mu_{\rm amb}}{\epsilon_{\rm amb}}}\left(\epsilon_{\rm amb}\,\mu_{\rm amb}-\xi_{\rm amb}^2\right)\,\underline{\underline{I}} & 2\xi_{\rm amb}\sqrt{\epsilon_{\rm amb}\,\mu_{\rm amb}}\,\underline{\underline{I}} \\ -2\xi_{\rm amb}\sqrt{\epsilon_{\rm amb}\,\mu_{\rm amb}}\,\underline{\underline{I}} & \sqrt{\dfrac{\epsilon_{\rm amb}}{\mu_{\rm amb}}}\left(\epsilon_{\rm amb}\,\mu_{\rm amb}-\xi_{\rm amb}^2\right)\,\underline{\underline{I}} \end{array}\right]\Bigg), \end{aligned} \tag{24}$$

after some straightforward manipulations.

2.3.2 Uniaxial dielectric ambient medium

Suppose that the ambient medium is a uniaxial dielectric medium. Without loss of generality, let us take the distinguished axis of this uniaxial medium to be aligned with the Cartesian z axis. Accordingly, its constitutive dyadic takes the form [19]

$$\underline{\underline{\mathbf{K}}}_{\rm amb} = \left[\begin{array}{cc} \left(\begin{array}{ccc} \epsilon_{\rm amb} & 0 & 0 \\ 0 & \epsilon_{\rm amb} & 0 \\ 0 & 0 & \epsilon_{\rm amb}^z \end{array}\right) & \underline{\underline{0}} \\ \underline{\underline{0}} & \mu_0\,\underline{\underline{I}} \end{array}\right]. \tag{25}$$

Within the present context of the SPFT, such an ambient medium arises in homogenization scenarios in which one of the constituent phases is a uniaxial dielectric medium and the other is either an isotropic dielectric medium or a uniaxial dielectric medium. Hence, we observe from eqs. (20) and (22) that only the upper three diagonal entries of $\underline{\underline{\mathbf{D}}}^+$, namely $\left[\underline{\underline{\mathbf{D}}}^+\right]_{nn}$ $(n=1,2,3)$, contribute to the estimate of the HCM constitutive dyadic. After some manipulations, these entries are evaluated

as

$$\begin{aligned}
\left[\underline{\underline{\mathbf{D}}}^{+}(\eta)\right]_{11} &= \left[\underline{\underline{\mathbf{D}}}^{+}(\eta)\right]_{22} \\
&= \frac{i\omega\mu_0\eta^2}{8}\left[\frac{1-\gamma_{\rm amb}}{\gamma_{\rm amb}} - \left(\frac{\epsilon^z_{\rm amb}}{\gamma_{\rm amb}\epsilon_{\rm amb}}\right)^2 \sqrt{\gamma_{\rm amb}}\,\tanh^{-1}\left(\sqrt{\gamma_{\rm amb}}\right)\right. \\
&\quad \left. + i\eta\frac{4\omega\left(3\epsilon_{\rm amb}+\epsilon^z_{\rm amb}\right)}{9}\sqrt{\frac{\mu_0}{\epsilon_{\rm amb}}}\right], \qquad (26)
\end{aligned}$$

$$\left[\underline{\underline{\mathbf{D}}}^{+}(\eta)\right]_{33} = \frac{i\omega\mu_0\eta^2}{4}\left\{\frac{1}{\gamma_{\rm amb}}\left[\frac{1+\gamma_{\rm amb}}{\gamma_{\rm amb}}\sqrt{\gamma_{\rm amb}}\;\tanh^{-1}\left(\sqrt{\gamma_{\rm amb}}\right)-1\right] + i\eta\frac{4\omega\sqrt{\epsilon_{\rm amb}\,\mu_0}}{9}\right\}, \qquad (27)$$

with the dimensionless scalar

$$\gamma_{\rm amb} = \frac{\epsilon_{\rm amb}-\epsilon^z_{\rm amb}}{\epsilon_{\rm amb}}. \qquad (28)$$

The representations (26) and (27) apply when $\gamma_{\rm amb}$ is complex–valued (with nonzero imaginary part). If $\gamma_{\rm amb}$ is real–valued then the representations (26) and (27) apply when $0 < \gamma_{\rm amb} < 1$; for $\gamma_{\rm amb} < 0$, the $\sqrt{\gamma_{\rm amb}}\;\tanh^{-1}\left(\sqrt{\gamma_{\rm amb}}\right)$ term in eqs. (26) and (27) should be replaced by $-\sqrt{-\gamma_{\rm amb}}\;\tan^{-1}\left(\sqrt{-\gamma_{\rm amb}}\right)$. In the scenario $\gamma_{\rm amb} > 1$ — which corresponds to nondissipative uniaxial dielectric ambient mediums with indefinite permittivity dyadics [34, 35, 36] — the components of $\underline{\underline{\mathbf{D}}}^{+}(\eta)$ are undefined, as are the corresponding components of $\underline{\underline{\mathbf{D}}}^{0}$ [20].

3 Discussion

The main results of this communication are the derivations of the eqs. (24), (26) and (27) which, when substituted into eq. (21), yield explicit formulations of the extended second–order SPFT for isotropic chiral HCMs and uniaxial dielectric HCMs (when supplemented with the corresponding expressions for $\underline{\underline{\mathbf{D}}}^{0}$ which are available elsewhere [19]). For more complex HCMs, numerical methods are needed to evaluate the depolarization dyadic $\underline{\underline{\mathbf{D}}}(\eta)$ and mass operator $\underline{\underline{\mathbf{\Sigma}}}^{[2]}(\eta, L)$.

It is helpful to consider the isotropic dielectric specialization of the extended second–order SPFT result (21). In this case, the constituent phases are both isotropic dielectric mediums with permittivities $\epsilon_{\rm a}$ and $\epsilon_{\rm b}$; in consonance, the ambient medium is also an isotropic dielectric medium with permittivity $\epsilon_{\rm amb}$. The corresponding second–order SPFT estimate of the HCM permittivity is

$$\epsilon^{[2]}_{\rm HCM} = \epsilon_{\rm amb} - \frac{1}{i\omega}\left(\frac{\Sigma^{[2]}(\eta, L)}{1+\Sigma^{[2]}(\eta, L)\,d(\eta)}\right), \qquad (29)$$

wherein the depolarization scalar may be expressed as the sum

$$d(\eta) = d^0 + d^+(\eta). \qquad (30)$$

The contribution to $d(\eta)$ associated with a vanishingly small depolarization region is provided by the well known result [19]

$$d^0 = \frac{1}{i3\omega\epsilon_{\rm amb}}, \qquad (31)$$

whereas the contribution associated with a depolarization region of nonzero volume may be extracted from eqs. (26) and (27) in the limit $\epsilon^z_{\rm amb} \to \epsilon_{\rm amb}$ as

$$d^+(\eta) = \frac{i\mu_0\omega\eta^2}{9}\,(3 + i2\eta\,\omega\sqrt{\epsilon_{\rm amb}\,\mu_0}\,)\,. \tag{32}$$

This expression for $d^+(\eta)$ also follows from eq. (24) when $\zeta_{\rm amb} = 0$ and $\mu_{\rm amb} = \mu_0$. The scalar mass operator term in eq. (29) is provided by the isotropic dielectric specialization of eq. (22) as

$$\Sigma^{[2]}(\eta, L) = f_{\rm a} f_{\rm b}\,[\chi_{\rm a}(\eta) - \chi_{\rm b}(\eta)]^2\,d^+(L), \tag{33}$$

with the polarizability density scalars

$$\chi_\ell(\eta) = -i\omega\left[\frac{\epsilon_\ell - \epsilon_{\rm amb}}{1 + i\omega\,(\epsilon_\ell - \epsilon_{\rm amb})\;d(\eta)}\right], \qquad (\ell = {\rm a,b}). \tag{34}$$

We note that the explicit expression for the extended second–order SPFT estimate of the HCM permittivity (29) is consistent with a corresponding result derived for the *unextended* SPFT, within the long wavelength limit represented by [4]

$$\epsilon^{[2]}_{\rm HCM} = \epsilon_{\rm amb} - \frac{1}{i\omega}\Sigma^{[2]}(L), \tag{35}$$

where $\Sigma^{[2]}(L) \equiv \Sigma^{[2]}(\eta, L)$ evaluated with d^0 in lieu of $d(\eta)$.

While our focus has primarily been on the extended second–order SPFT, we bear in mind that the expressions (24), (26) and (27) can also be deployed in extended versions of other homogenization formalisms, such as the frequently used Bruggeman and Maxwell Garnett formalisms. In fact, the Bruggeman homogenization formalism is equivalent to the zeroth order SPFT [9]. Hence the extended Bruggeman estimate of the HCM constitutive dyadic is simply the dyadic $\underline{\underline{\mathbf{K}}}_{\rm amb}$ which may be extracted from eq. (19). Like the Bruggeman formalism, the Maxwell Garnett formalism is based on depolarization dyadics [24], but with one of the constituent phases playing the role of the ambient medium. Therefore, the Maxwell Garnett formalism — including its incremental [37] and differential [38] variants — may be extended by implementing the appropriate η–dependent depolarization dyadic $\underline{\underline{\mathbf{D}}}(\eta)$.

We close by considering the question: For what range of constituent particle sizes are the extended homogenization formalisms discussed herein appropriate? An upper bound is straightforwardly established by the requirement that the particles must be small relative to the electromagnetic wavelength(s), in order to be consistent with the notion of homogenization. For optical wavelengths, the linear dimensions of the constituent particles must therefore be at most 38–78 nm. A lower bound comes into effect because, at sufficiently small length scales, quantum processes cannot be neglected in the description of the constituent particles and their interactions. Accordingly, the lower bound is material–dependent. We note that for very small constituent particles, their constitutive parameters may differ significantly from those of the corresponding bulk materials and depend upon the shape and size of the particles [39]. This is particularly the case for metallic particles smaller than the mean free path of conduction electrons in the bulk metal, wherein the mean free path may be dominated by collisions at the particle boundary [40]. Within this particle–size regime, a recent study using spectroscopic ellipsometry demonstrated that an extended Maxwell Garnett homogenization formalism adequately characterizes a HCM based on silver nanoparticles as small as 2.3 nm [41]. This study also highlights the prospects of implementing extended homogenization formalisms, such as those described herein, in particle sizing applications for nanocomposites.

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